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*Studies in feldspar equilibria at the Geophysical Laboratory,  
Washington.*

By O. F. TUTTLE.

[Abstract of lecture, May 16, 1951.]

EVIDENCE for the existence of four series of alkali-feldspars has been obtained from X-ray studies of over forty analysed feldspars.<sup>1</sup> Optical studies confirm these findings, but suggest that transitional forms may occur. The four series are: high-sanidine-high-albite; sanidine-high-albite; orthoclase-low-albite; microcline-low-albite. High-sanidine has the optic axial plane parallel to (010) in the potash-rich members, whereas it is perpendicular to (010) in sanidine.

Synthetic feldspars crystallized at the liquidus and natural feldspars heated for prolonged periods at high temperatures belong to the high-sanidine-high-albite series. The variation of optical properties of sanidinized potash-rich feldspars studied by Edmondson Spencer<sup>2</sup> has been found to extend, with no evidence of a discontinuity, through the soda-rich compositions to high-albite. Prolonged heating of the synthetic material below the solvus (i.e. below 660° C. for compositions near  $\text{Ab}_{50}\text{Or}_{50}$ ) results in unmixing into two phases, giving a synthetic cryptoperthite.

Nearly all alkali-feldspar phenocrysts from extrusive rocks fall into the sanidine-high-albite series, together with feldspars from certain high-temperature pegmatites. X-ray studies of these feldspars show that most specimens have unmixed to a submicroscopic perthite composed of nearly pure potash-feldspar (sanidine) and pure albite. These cryptoperthites can be homogenized by heating for a short time at 900° C. Most of the specimens examined are optically monoclinic before heating.

<sup>1</sup> N. L. Bowen and O. F. Tuttle, unpublished. [cf. M.A. 11-325, 327.]

<sup>2</sup> E. Spencer, The potash-soda-feldspars. I. Thermal stability. Min. Mag., 1937, vol. 24, pp. 453-494.

This optically monoclinic character of a cryptoperthite composed of triclinic albite and monoclinic sanidine is apparently due to submicroscopic twinning of the albite phase, as has been established by C. Oftedahl<sup>1</sup> for cryptoperthites composed of orthoclase and low-albite. Homogenization of these cryptoperthites with compositions between  $Ab_{67}$  and  $Ab_{100}$  causes them to become optically triclinic, whereas compositions between  $Ab_{57}$  and  $Ab_0$  remain monoclinic.

The orthoclase-albite series was recognized and described in detail by Edmondson Spencer (*loc. cit.*). These are also unmixed, in this case to orthoclase and low-albite. X-ray and optical studies of heat-treated material suggest that compositions rich in potash can be readily homogenized with little change in optical properties, but compositions near  $Ab_{50}Or_{50}$  resist homogenization and it appears that such specimens must be sanidinized before they will become homogeneous.

The microcline-albite series is less well known as only five analysed specimens are available for study. In all cases submicroscopic albite is present which can be 'dissolved' on heating at 900° C. for a short time.

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<sup>1</sup> C. Oftedahl, Studies on the igneous rock complex of the Oslo region. IX. The feldspars. *Skrifter Norske Vidensk.-Akad. Oslo, I. Mat.-Naturv. Kl.*, [1949], for 1948, no. 3, 71 pp. [M.A. 11-11.]

*X-ray determination of centrosymmetry in three feldspars.*<sup>1</sup>

By S. W. BAILEY, R. B. FERGUSON, and W. H. TAYLOR.

Crystallographic Laboratory, Cavendish Laboratory, Cambridge.

[Read May 16, 1951.]

*Introduction.*

THE recently developed X-ray methods for determining the presence or absence of certain symmetry elements in a crystal have been applied, with reference to centrosymmetry, to orthoclase, sanidine, and albite. The methods depend upon the fact that the presence or absence of certain symmetry elements in a structure affects the probability distribution of the X-ray intensities. The methods were first investigated theoretically by Wilson (1949), and have since been extended and applied by Wilson and several of his colleagues. The potential importance of these new methods for problems of mineralogical interest justifies the brief descriptions of the three methods of testing for centrosymmetry given below; further details may be had from the original papers to which we refer. The theory assumes that a relatively large number of atoms occupy general positions in the unit cell, and that the intensity distribution is not dominated by a few of them; in practice the methods tolerate considerable departure from these conditions and may be applied to a wide variety of crystals.

*Methods.*

The presence of a symmetry-centre tends to create sharp intensity differences from reflection to reflection; the absence of one tends to create uniformity in the intensity distribution. The effect is sometimes apparent on inspection of single-crystal photographs, but it may always be investigated statistically by considering the accurately measured intensities of a large number of general reflections, *hkl*. Special reflections (*hk0*, *0kl*, &c.) may not be sufficient for a satisfactory test for centrosymmetry in general, because a zone may be centrosymmetric when the crystal as a whole is not.

<sup>1</sup> The substance of this paper was presented briefly during discussion at a special meeting of the Mineralogical Society devoted to feldspars, May 16, 1951.

1.  $N(z)$  test (Howells, Phillips, and Rogers, 1950).—The reflections are divided into several groups containing approximately equal numbers of reflections. Each group consists of all reflections, including those

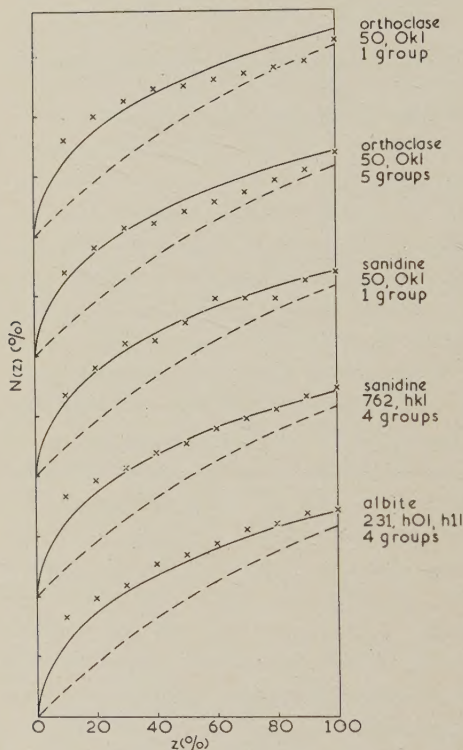


FIG. 1.  $N(z)$  plots of the X-ray intensity distributions for orthoclase, sanidine, and albite. The crosses represent experimental values. The solid line curves give the theoretical values expected for centrosymmetric crystals ( $\bar{1}$ ); the broken line curves give the theoretical values for non-centrosymmetric crystals (1). Individual curves have been displaced vertically 40% for clarity.

accidentally absent, within a small range of  $\sin \theta$  or  $(\sin \theta / \lambda)^2$ . For each group the average intensity  $\langle I \rangle$  is calculated, and the percentage  $N(z)$  of the total number of reflections is found for which the intensity is less than or equal to progressive fractions  $z$  of the average intensity. Finally, for each  $z$  the weighted mean of  $N(z)$  over all  $\sin \theta$  groups is compared graphically with the theoretical values. In the accompanying figure (fig. 1), theoretical values for the centrosymmetric case ( $\bar{1}$ ) are shown as solid curves, for the non-centrosymmetric (1) as broken curves.

2. Wilson ratio test (Wilson, 1949; Howells, Phillips, and Rogers, 1950).—The reflections may be divided into several  $\sin \theta$  groups as in the  $N(z)$  test or they may be treated as one large group. The average structure amplitude

and the average intensity  $\langle I \rangle$  are determined for each group, and then the ratio  $\langle |F|^2 \rangle / \langle I \rangle$  is calculated. The weighted mean over all  $\sin \theta$  groups of this expression, known as the Wilson ratio  $\rho$ , has the theoretical values 0.637 for ( $\bar{1}$ ) and 0.785 for (1).

3. Variance test (Wilson, 1951).—Again a distinguishing ratio is deter-

mined, in this case  $\langle(I-\langle I \rangle)^2\rangle/\langle I \rangle^2$ , the mean square deviation of  $I$  from its average. This ratio has the theoretical values 2.00 for  $(\bar{I})$  and 1.00 for (1).

### Material and results.

*Orthoclase*.—We have applied all three methods to 50  $0kl$  intensities published by Chao, Hargreaves, and Taylor (1940) for a typical orthoclase from Mogok in Upper Burma, specimen *C* of Edmondson Spencer's suite of potash-soda-feldspars (Spencer, 1930, 1937). Cole, Sörum, and Kennard (1949) showed that these  $0kl$  intensities are somewhat inaccurate in that some reflections which are actually present were listed with zero intensity. We have therefore altered the published intensity data by adopting the calculated intensities for these few reflections.

The  $N(z)$  plots are shown in the figure both for a single grouping of the reflections and for a division into five overlapping groups, this division simply having the effect of smoothing the plot. The plots are only in fair agreement with the theoretical curve for centrosymmetry. Howells, Phillips, and Rogers (1950) discuss the factors which may alter the shape of the plot. In particular, extinction frequently reduces the intensities of the strongest reflections, and the plot may agree with the theoretical curve at the low intensity end where extinction is not appreciable, but it may sag as the intensity increases. This may well explain the shape of the plots for orthoclase. Although our  $N(z)$  plot is not convincing evidence for a centre of symmetry in orthoclase, the Wilson ratio of 0.626 (theoretical for  $(\bar{I})$ , 0.637) and the variance average of 2.12 (theoretical for  $(\bar{I})$ , 2.00) shown in table I leave little doubt that orthoclase is

TABLE I. Wilson ratios and variance averages of three feldspars.

|                                | Number<br>of<br>reflections. | Type of<br>reflections. | Limits of<br>( $\sin \theta/\lambda$ ) <sup>2</sup> . | Number<br>of<br>groups. | Wilson<br>Ratio<br>$\langle F ^2\rangle/\langle I \rangle$ . | Variance<br>average<br>$\langle(I-\langle I \rangle)^2\rangle/\langle I \rangle^2$ . |
|--------------------------------|------------------------------|-------------------------|-------------------------------------------------------|-------------------------|--------------------------------------------------------------|--------------------------------------------------------------------------------------|
| Orthoclase ..                  | 50                           | $0kl$                   | 0.025–0.400                                           | 1                       | 0.626                                                        | 2.12                                                                                 |
| Sanidine ..                    | 50                           | $0kl$                   | 0.025–0.400                                           | 1                       | 0.664                                                        | 2.54                                                                                 |
| Sanidine ..                    | 762                          | $hkl$                   | 0.025–0.400                                           | 4                       | 0.561                                                        | —                                                                                    |
| Albite ..                      | 231                          | $h0l, h1l$              | 0.025–0.500                                           | 4                       | 0.564                                                        | —                                                                                    |
| Theoretical values $(\bar{I})$ |                              |                         |                                                       |                         | 0.637                                                        | 2.00                                                                                 |
| (1)                            |                              |                         |                                                       |                         | 0.785                                                        | 1.00                                                                                 |

centrosymmetric. A larger number of reflections such as we describe below for sanidine and albite would have been more favourable for statistical averaging and might have improved our  $N(z)$  plot.

*Sanidine*.—The intensities we have used are 762 of those obtained for the high accuracy analysis of sanidine reported by Cole, Sörum, and

Kennard (1949); the intensities were taken from Cole (1949). This sanidine is the material from Upper Burma referred to above, but sanidinized by heating at 1070° C. for 300 hours (Spencer, 1937). For comparison with our results for orthoclase, we have analysed the same 50 *Ok*l reflections of sanidine in one group by all three methods. The  $N(z)$  curve agrees fairly well with the theoretical curve for ( $\bar{1}$ ), and the Wilson ratio and the variance average agree reasonably well with the theoretical values for centrosymmetry. There is an apparent discrepancy between the Wilson ratio and the variance average: the former, 0.664, departs from the centrosymmetric value towards the non-centrosymmetric value of 0.785, whereas the latter, 2.54, shows a departure in the opposite direction, i.e. away from the non-centrosymmetric value of 1.00. This is explained by the fact that all sources of error in the variance test tend to increase the experimental value, whereas various sources of error affect the Wilson ratio differently.

When all 762 reflections are used for sanidine the  $N(z)$  plot agrees very closely with the theoretical curve for the centrosymmetric case. The Wilson ratio of 0.561 is appreciably lower than the theoretical value for ( $\bar{1}$ ), 0.637. The reason for this deviation is not known.

*Albite*.—We have used a very pure typical low-temperature albite from a pegmatite located at Ramona, California. We are indebted to Dr. W. T. Schaller and Dr. C. S. Ross of the U.S. Geological Survey for the use of this specimen. The material is part of a suite of plagioclase feldspars kindly supplied by Professor R. C. Emmons of the University of Wisconsin, who is presently to publish chemical, optical, and petrological descriptions of them. An X-ray investigation of several of these specimens carried out in this Laboratory has already been published (Cole, Sörum, and Taylor, 1951). This albite from California is at present being analysed in detail by three-dimensional Fourier methods by one of us (R. B. F.) in order to refine the earlier parameters published by Taylor, Darbyshire, and Strunz (1934). The results of our statistical analysis carried out on 231 reflections of the types *h*0*l*, *h*1*l*, prove conclusively that albite is centrosymmetric. The low value of the Wilson ratio, 0.564, is surprisingly close to our value of 0.561 for the 762 reflections of sanidine.

#### *Discussion and conclusions.*

This newly developed X-ray method of testing certain types of crystals for centrosymmetry (and, indeed, for other symmetry elements, see Wilson (1950) and Rogers (1950)), should prove a very important addi-

tion to the older morphological, etching, and piezo- and pyro-electric methods. If a crystal satisfies, or nearly satisfies, the conditions mentioned earlier, the presence or absence of specified symmetry elements (in particular, symmetry-centres) can probably be decided. Because the method requires only the minute fragment necessary for single-crystal photographs, or in favourable cases sufficient powder for a powder photograph, it is a particularly powerful tool for the structural mineralogist. The intensity data used for the test are those normally prepared for structure analysis, and so the test may be quickly applied as a routine part of any structure analysis in which the space-group is not uniquely determined by the ordinary methods.

In relation to the structures of the feldspars, the method removes one of the previous uncertainties, that concerning the space-group, and our present work has shown that the assumption of the holohedral space-group  $C2/m$  for orthoclase and sanidine by Chao, Hargreaves, and Taylor (1940) and later by Cole, Sörum, and Kennard (1949) was correct. It is doubtful whether the method will prove of direct assistance in clarifying the Si-Al order-disorder phenomena widely believed to be primarily responsible for the polymorphism of the alkali-feldspars. This X-ray test for centrosymmetry depends upon the diffracting powers of the atoms and thus, for the purpose of the test, the Si and Al atoms will behave almost identically. If one or more of the feldspars should have a structural arrangement in which the Si and Al atoms are segregated into positions compatible with only a hemihedral space-group, it is almost certain that the statistical method would show the crystal to be centrosymmetric if all the other atoms were arranged according to the corresponding holohedral space-group.

*Acknowledgements.*—We wish to express our thanks to Professor Sir W. Lawrence Bragg for his interest in this problem, and to E. R. Howells, who has kindly given us much helpful advice. One of us (S. W. B.) is indebted to the U.S. Government for the grant of a Fulbright Scholarship, and another (R. B. F.) to the National Research Council of Canada for the grant of a Post-Doctorate Overseas Fellowship.

*Summary.*—Three statistical tests, depending upon the probability distribution of X-ray intensities, have been applied to orthoclase, sanidine, and albite. It is concluded that these three feldspars possess centres of symmetry, justifying the assumption of holohedral symmetry made in earlier structure analyses.

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*Taaffeite, a new beryllium mineral, found as a cut gemstone.*<sup>1</sup>

By B. W. ANDERSON, B.Sc., F.C.S., C. J. PAYNE, B.Sc.

Laboratory of the Diamond, Pearl, and Precious Stone Trade  
Section of the London Chamber of Commerce.

and G. F. CLARINGBULL, B.Sc., Ph.D., F.G.S.

With microchemical analysis by M. H. HEY, M.A., D.Sc.

Department of Mineralogy, British Museum.

[Read June 7, 1951.]

IN October 1945 Count Taaffe,<sup>2</sup> a brilliant if unorthodox Dublin gemmologist, in the course of examining a motley collection of gemstones, came across a small mauve stone which puzzled him greatly. The stone had the appearance, and most of the characters, of spinel, but afforded clear evidence of double refraction. As recounted below, this stone was later found to belong to an entirely new mineral species—the only case hitherto known where a mineral has been first encountered as a faceted gem.

Since the precise circumstances of such a discovery have both human and technical interest, it seemed best to obtain from Count Taaffe his own account of the event. This is accordingly given below before proceeding to the more formal presentation of the data on the new mineral, which has been named taaffeite in honour of its discoverer.

On one of my rounds in search of gems I came to Mr. Robert Dobbie, watchmaker and working jeweller in Fleet Street, Dublin; he allowed me in his genial way to go through all his boxes in which he kept stones, to pick out any that were real—most of them were glass—and to make him an offer for them. He only needed the pastes for jobbing. That was one day in October 1945. I started right away in his workshop where he had cleared part of his bench and I sorted out what looked like stones, one by one. Most of the stones and pastes were crudely divided according to colour. The pastes were mostly new, but the stones were without exception broken out of jewellery, badly rubbed or chipped in many cases. They had been in the workshop since the times of Mr. Robert Dobbie's father, some for twenty years.

<sup>1</sup> Preliminary accounts appeared in *Nature*, London, 1951, vol. 167, p. 438; *Gemmologist*, 1951, vol. 20, pp. 75–77 (p. 76, 'Taaffeite', Anderson); *Journ. Gemmology*, 1951, vol. 3, pp. 75–77. [M.A. 11–309.]

<sup>2</sup> Count Edward Charles Richard Taaffe, born in Bohemia 1898, only son of late Henry, Count Taaffe (12th Viscount Taaffe of Corran, Baron of Ballymote, Co. Sligo) and of Magda, Countess Taaffe.

I found there blue zircons, opals, garnets (even demantoids), citrines, amethysts, spinels, very few Siam rubies, some poor emeralds and sapphires, in fact stones of the usual mixture as they accumulate with working jewellers—their origin as manifold as their appearance. After several hours I showed Mr. Dobbie what I had selected so far; maybe there were 50 or 100 stones. I paid and arranged to continue the next day, and took the 'bag' home. I continued till I had gone through every stone in the house; it took two or three days I think. The total purchase price was £14; it was very easy to come to terms with Mr. Dobbie.

Now, at home, I washed the stones, which were very dirty, and started sorting them and testing each one of them, because I am always afraid of a wrong identification if I go only by appearance. First I sorted according to colour. The taaffeite was put into the little box containing violet and lilac stones; then viewed under my Bausch and Lomb binocular Greenough microscope, without stage, on white paper, with illumination from above, using a flexible table-lamp with tin reflector, ordinary 100-watt bulb with a flexible lens-holder as condenser. Magnification is  $21\times$  (objective  $1.4\times$ , ocular  $15\times$ ). Holding each stone in tweezers, and viewing it under different angles and from several sides, I look for double refraction and evidence of the stone's identity, e.g., cracks in the surface on opposite 'poles' denoting corundum, &c. Mind you, I do that with every stone, and only in the rarest cases have I to apply other methods to see whether the stone is doubly refractive or not. One of the stones in which I can sometimes not detect double refraction is topaz; I do not understand why. I have to twist and twist and look into every facet until I can see it. But not so with the taaffeite. In a certain direction every speck of dust on the back and every scratch appeared double like on a badly wobbled snapshot.

Next step: specific gravity. Sinking in bromoform. Sinking in methylene iodide, but more slowly than corundum. Clerici solution was unobtainable then. My only balance was a pocket diamond balance which I had to hold by the tassel. As B. W. Anderson taught me in his book 'Gem testing', I determined the S.G. by hydrostatic weighing in water, holding the tassel and repeating the observation ten times. The average of ten results was 3.62.

Facets were scarcely visible in methylene iodide, so I gathered that the refractive index was near spinel. Refractometer not obtainable. But the double refraction? I checked it with crossed nicols and got normal extinction at 90 degrees. I became puzzled. I could see no bands in the spectrum. The two spectroscopes I had at the time were not very good and the lighting system quite inadequate. I panicked, sent the stone to B. W. Anderson at the Laboratory of the London Chamber of Commerce, and begged for help. I wrote on November 1 1945: 'This time a new riddle: what is this mauve stone? It seems to me to answer all characteristics of spinel, yet it shows double refraction: doubling of facets visible under the Greenough, extinction when polarized, though with queer colour effects. Could anomalous double refraction be so strong? R.I. too high for topaz, S.G. too low for corundum. What is it?'

B. W. Anderson replied on November 5: 'You have sent a *most* interesting stone in that mauve spinel (I call it that on the basis of its absorption spectrum: my colleague is so horrified at its anomalous optics that he doubts whether it is spinel). The indices are 1.717 and 1.721, clear double refraction, giving a clear uniaxial interference figure through the table facet. The stone is so remarkable that I should like your permission to have an X-ray analysis made, if possible without harming the stone. I should also like to write it up for the *Gemmologist*, giving full credit to you for discovering it. Would you agree?'

*Optical and density determinations (B. W. A. and C. J. P.).*

It will be seen from the above account how an acute, experienced, and painstaking observer can obtain most of the data needed to identify gemstones with the simplest of apparatus; even without the aid of a refractometer, which is rightly regarded as 'essential' by most gemmologists.

The mauve-coloured stone when it arrived weighed 1.419 carats. Its shape suggested that probably it had been cut in Ceylon. The refractive indices given in the first reply to Taaffe were not quite accurate. A little later quite good readings for the ordinary ray were obtained by the minimum deviation method using a prism angle (provided by the facets) of  $48^\circ$ . The indices were determined for lithium (6708 Å.) light and for Fraunhofer's g line (4226 Å.), as well as for sodium light, in order to measure the dispersion of the stone. The results were:

|           |   |   |        |        |         |
|-----------|---|---|--------|--------|---------|
| $\lambda$ | . | . | 6708   | 5893   | 4226 Å. |
| $\omega$  | . | . | 1.7191 | 1.7230 | 1.7390  |

This gave a Li-g dispersion figure of 0.0199, corresponding to approximately 0.019 (for the standard B-G range). This compares quite closely with the dispersion of spinel, which is 0.0205 to 0.0207 for stones having a refractive index of 1.715 for sodium light.<sup>1</sup> Later still, an opportunity arose to measure both refractive indices on a Zeiss Abbe-Pulfrich refractometer, which gave the following figures:  $\omega$  1.7230,  $\epsilon$  1.7182,  $\omega - \epsilon$  0.00475.

The density, as determined by hydrostatic weighing in ethylene dibromide, was measured as 3.621 at  $15^\circ$  C. Later, a more accurate estimate was achieved by finding blue spinel pebbles which matched the taaffeite closely in density as evidenced by their behaviour in Clerici solution. These were of sufficient size to enable reliable determinations to be made by hydrostatic weighing in ethylene dibromide. By choosing one specimen which was slightly denser and the other slightly less dense than the taaffeite, limiting values were obtained. On the basis of these experiments  $3.613 \pm 0.002$  emerges as the most probable figure.

The absorption spectrum was very weak, but bore a distinct resemblance to that of blue spinel, particularly two weak bands in the green at 5530 and 5580 Å., corresponding bands in spinel being 5520 and 5570 Å., which is within the limits of error for such diffuse bands. The strongest bands in blue spinel, a broad absorption centred near 4590 Å. and a narrower band at 4800 Å. are replaced by a very weak narrowish

<sup>1</sup> B. W. Anderson and C. J. Payne, *Min. Mag.*, 1937, vol. 24, p. 548.

band at about 4750 and a diffuse region centred near 4600 Å. Recently, a photograph taken on a small diffraction grating spectograph revealed the unexpected presence of a narrow and fairly intense band in the near ultra-violet at approximately 3820 Å., entirely absent in a corresponding spectrogram of blue spinel.

To enable X-ray and chemical work to be carried out, Count Taaffe generously assented to portions of the stone being removed. It will be realized that this was a crucial business since at that time there was only this one specimen of the mineral known. The work was skilfully carried out by Charles Mathews (Lapidaries) Ltd., in two stages, reducing the stone first to 0.95 carat, and then 0.56 carat which is its present weight. The stone was refaceted and polished in order to preserve its appearance as an attractive gemstone.

A summary of the properties of Taaffe's original specimen can now be given:

| $\epsilon$ . | $\omega$ . | $\omega - \epsilon$ . | Density. | Hardness. |
|--------------|------------|-----------------------|----------|-----------|
| 1.7182       | 1.7230     | 0.00475               | 3.613    | 8         |

It was not until October 1949 that a second specimen was found. This stone weighed 0.87 carat and was of a very pale mauve colour, in fact it was almost colourless. The stone was sent to the Laboratory in Hatton Garden together with a hundred other stones in a case (presumably belonging to a collector). They were sent in to be tested in the ordinary way of business by a Hatton Garden merchant. Not much information about the origin of the stones could be gleaned from the nature of the collection. It was obvious that none of the stones had been cut in Asia: though not very well cut or polished they had regular facets and were all symmetrical in styles which are included in the general term 'mixed cut'. The collection consisted mostly of green sapphires, though one of them was found to be a Ceylon kornerupine (Ceylon was probably also the source of the sapphires), and spinels many of which were small, pale-coloured stones. These spinels were very typical, having  $n_D$  1.715. They were tested on a refractometer which was in a bad condition, as owing to external circumstances no other instrument was available at the time. One stone, however, had  $n_D$  1.721 or 1.720: it was impossible to see birefringence owing to the small and badly polished table facet and the worn surface of the refractometer glass. It did, however, show a uniaxial negative interference figure of a size which suggested a small birefringence. The tester (C. J. P.) then realized that he had found a second taaffeite. The birefringence was confirmed when other refractometers were available, and finally the stone was sent to a lapidary to

have the table facet enlarged, well polished, and truly plane. More elaborate optical work was done and the stone was found to have the following properties:

| $\epsilon$ . | $\omega$ . | $\omega - \epsilon$ . | Density. | Hardness. |
|--------------|------------|-----------------------|----------|-----------|
| 1.7167       | 1.7208     | 0.00412               | 3.60     | 8         |

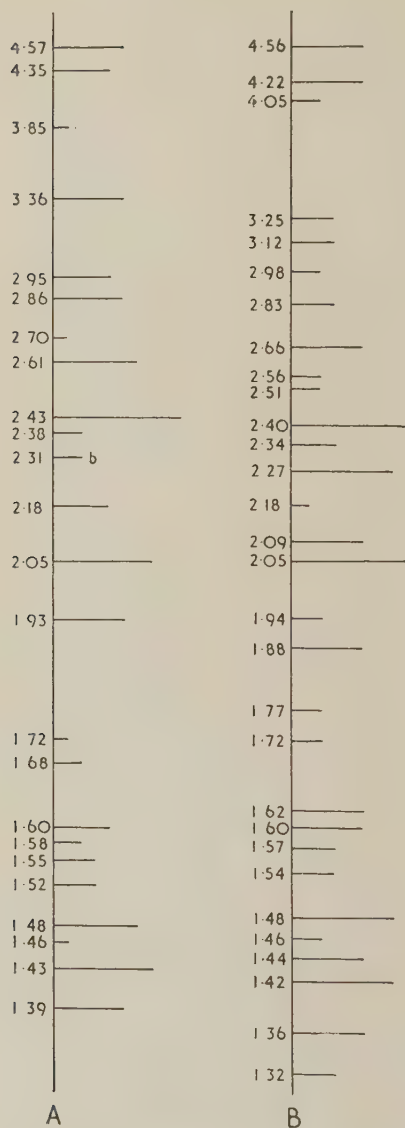
It was confirmed by X-ray analysis that this stone was of the same species as that of Count Taaffe.

The hardness of the first taaffeite was cautiously tested by trying its edge against polished surfaces of quartz and topaz, and was found to be nearly equal to the latter. On the second specimen, confirmation of this was kindly provided by Dr. W. Stern of the Diamond Information Bureau, who carried out micro-indentation tests with a double cone indenter, and found the hardness of taaffeite by this method to be slightly greater than natural spinel and slightly less than synthetic spinel, when these were tested at the same time and under the same conditions.

Before passing on to a discussion of the composition and structure of the mineral, there is one more property that may be mentioned: its luminescence. Under ultra-violet radiation of both 2536 Å. and 3650 Å. the stones remained inert; but under X-rays both showed a distinct green fluorescence. In Taaffe's stone this was not nearly so brilliant as in the second stone, presumably due to the presence of rather more iron in its composition, to which the enhancement in colour, density, and refractive indices may also reasonably be ascribed. The X-ray luminescence might provide a rapid test for the mineral were it not that spinel of similar appearance gives an almost identical green glow under the rays.

*X-ray determinations (G. F. C.) and chemical analysis (M. H. H.).*

A preliminary examination by X-ray methods was made of Count Taaffe's stone as originally submitted for testing; this confirmed the optical indications that it could not be spinel. It was therefore decided to make a more detailed study and for this a small portion sawn from the region of the culet was used. The fragment so obtained was lightly crushed in a diamond mortar and one of the resulting smaller fragments powdered and photographed. The measurements obtained (table I and fig. 1A) were used to search for comparable data in the literature and in the British Museum records of unpublished X-ray photographs. No such data were traced. The powder spacings were subsequently indexed by first determining the  $l$  indices by the superposition of a powder photograph upon a  $c$ -axis rotation photograph taken in the same camera and



then determining the remaining indices by reference to the appropriate Bunn chart. A

TABLE I. Powder data for taafeite. (Film no. X 3559b.)

| <i>hkl</i>         | <i>I</i> | <i>d</i> |
|--------------------|----------|----------|
| 0004               | m        | 4.57     |
| 10 $\bar{1}$ 2     | mw       | 4.35     |
| 10 $\bar{1}$ 3     | vvw      | 3.85     |
| 10 $\bar{1}$ 4     | m        | 3.36     |
| 10 $\bar{1}$ 5     | mw       | 2.95     |
| 11 $\bar{2}$ 0     | m        | 2.86     |
| 11 $\bar{2}$ 2     | vvw      | 2.70     |
| 10 $\bar{1}$ 6     | ms       | 2.61     |
| 11 $\bar{2}$ 4     | vvs      | 2.43     |
| 20 $\bar{2}$ 2     | vw       | 2.38     |
| 20 $\bar{2}$ 3     | vw b     | 2.31     |
| 10 $\bar{1}$ 7     |          |          |
| 20 $\bar{2}$ 4     | mw       | 2.18     |
| 20 $\bar{2}$ 5     | s        | 2.05     |
| 20 $\bar{2}$ 6     | m        | 1.925    |
| 20 $\bar{2}$ 8     | vvw      | 1.720    |
| 21 $\bar{3}$ 5     | vw       | 1.684    |
| 21 $\bar{3}$ 6     | mw       | 1.596    |
| 20 $\bar{2}$ 9     | vw       | 1.576    |
| 30 $\bar{3}$ 4     | w        | 1.554    |
| 21 $\bar{3}$ 7     | w        | 1.522    |
| 2.0. $\bar{2}$ .10 | ms       | 1.476    |
|                    | vvw      | 1.455    |
| 22 $\bar{4}$ 0     | s        | 1.428    |
| 2.0. $\bar{2}$ .11 | m        | 1.387    |

Strongest lines 2.43, 2.05, 1.43.

6 cm. diam. camera, Cu-K $\alpha$   
( $\lambda = 1.542\text{\AA}$ .) radiation.

Intensity scale in decreasing order:  
vvs, vs, s, ms, m, mw, w, vw, vvw;  
b, broad.

second chip was set up to rotate about the optic (*c*) axis and a rotation photograph in this position gave a lattice spacing

FIG. 1. Spacing-intensity plots of X-ray powder data of: (A) taafeite. (B) artificial compound presumed to be  $\text{MgO} \cdot \text{BeO} \cdot 2\text{Al}_2\text{O}_3$  (H. F. McMurdie in R. F. Geller et al., 1946).

The *d*-spacings are plotted as reciprocals and their approximate values in Ångström units are given against each line, the length of which is an indication of intensity. b = broad.

along the axis of  $18.38 \text{ \AA}$ . The chip was then set up by trial about another axis normal to the first (and subsequently found to be  $b$ ) and a Laue photograph taken along the optic axial direction. The dihexagonal ( $6mm$ ) symmetry of this is clearly seen in fig. 2, and serves to distinguish

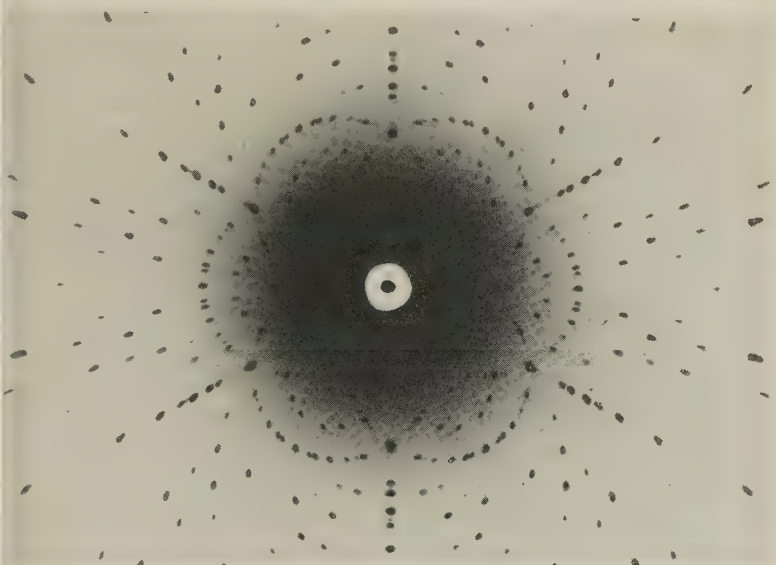


FIG. 2. X-ray Laue photograph of taafeite along the optic ( $c$ ) axis. Crystal plate distance  $3.5 \text{ cm}$ . Tungsten radiation.

the mineral from spinel, the only possible comparable axis of which is ditrigonal. Measurement of the  $b$  rotation and sets of oscillation photographs about  $b$  and  $c$  axes gave  $a \ 5.72 \text{ \AA}$ . The diffraction symmetry from the Laue and oscillation photographs is  $6/mmm$ , and this, together with the absence of all odd orders of  $(000l)$ , leads to the unequivocal space-group  $D_6^6 - C6_32$ . The axial ratio  $c/a$  is  $3.21$ .

Still another small chip was used to obtain a spectrogram in which the following elements were detected: Mg, Al, Be.

For the microchemical analysis reported below further pieces totalling about  $12 \text{ mg}$ . had been cut from the original stone. About half of this material was used for a preliminary run and the remaining  $6.16 \text{ mg}$ . for the analysis proper.

The atomic contents of the unit cell are derived from the analysis and cell dimensions ( $a \ 5.72 \text{ \AA}$ .,  $c \ 18.38 \text{ \AA}$ .) with the observed density of  $3.613$ .

The iron is assumed to be all ferric because there is an excess of divalent ions without it; the colour of the mineral is also appropriate to ferric ions.

TABLE II. Microchemical analysis and unit-cell contents of taaffeite.

|                                |   |   |   |       |       |   |   |   |      |        |
|--------------------------------|---|---|---|-------|-------|---|---|---|------|--------|
| Al <sub>2</sub> O <sub>3</sub> | . | . | . | 70.0  | Al    | . | . | . | 15.6 | } 16.4 |
| Fe <sub>2</sub> O <sub>3</sub> | . | . | . | 5.9   | Fe''' | . | . | . | 0.8  |        |
| MgO                            | . | . | . | 13.4  | Mg    | . | . | . | 3.8  | } 8.8  |
| BeO                            | . | . | . | 11.0  | Be    | . | . | . | 5.0  |        |
| Analyst, M. H. Hey             |   |   |   | 100.3 | O     | . | . | . | 33.4 |        |

The figure for beryllia is thought to be somewhat high. Owing to the small amount of material analysed, the unit-cell content can only be approximate and the cell formula is given in whole numbers. Taaffeite, in which some aluminium is replaced by ferric iron, may thus be regarded as intermediate between spinel and chrysoberyl and have the ideal formula  $\text{Be}_4\text{Mg}_4\text{Al}_{16}\text{O}_{32}$ ; it appears to be the only known mineral having both essential Be and Mg.

Assuming an approximately close-packed oxygen framework for the structure it is evident from a consideration of the cell dimensions that this framework will be built of eight close-packed oxygen sheets in hexagonal (i.e., ABAB) stacking, the distance between the centres of adjacent sheets being  $18.38/8 = 2.30 \text{ \AA}$ . The distance between centres of adjacent oxygens in the sheets is found from the  $a$  lattice-dimension to be  $5.72/2 = 2.86 \text{ \AA}$ . These figures agree closely with the corresponding dimensions for spinel<sup>1</sup> in which the oxygen sheets in cubic (i.e., ABCABC) stacking are  $2.32 \text{ kX}$  apart and the atoms within the sheets  $2.85 \text{ kX}$  between centres. As might be expected therefore the two structures appear to be closely related, with the hexagonal axis of taaffeite corresponding to a trigonal axis of spinel. A more detailed examination of the structure of taaffeite is in progress.

The stone found by Payne in 1949 was checked by taking a randomly oriented rotation photograph through the tip of the pavilion and superposing it on the powder photograph of Taaffe's stone.

We are indebted to Dr. J. B. Nelson for drawing our attention to the description of an artificial compound of the same essential composition and having closely related physical properties.<sup>2</sup> The published X-ray powder spacings for this substance stated to be 'diffraction data from patterns for the dominant phase in compositions  $\text{MgO} \cdot \text{BeO} \cdot 2\text{Al}_2\text{O}_3$  and  $\text{MgO} \cdot 2\text{BeO} \cdot 2\text{Al}_2\text{O}_3$ ' are not, however, in good agreement with those for taaffeite although they show a rough resemblance (fig. 1B).

<sup>1</sup> W. L. Bragg, The atomic structure of minerals. 1937, p. 94. [M.A. 7-7.]

<sup>2</sup> R. F. Geller, P. J. Yavorsky, B. L. Steierman, and A. S. Creamer. Journal of Research of the National Bureau of Standards, 1946, vol. 36, pp. 277-312 (Research Paper R.P. 1703). [M.A. 11-369.]

*The amygdale minerals in the Tertiary lavas of Ireland.*  
*I. The distribution of chabazite habits and zeolites in the*  
*Garron plateau area, County Antrim.*

By GEORGE P. L. WALKER, M.Sc., F.G.S.

Department of Geology, University of Leeds.

[Read June 7, 1951.]

1. INTRODUCTION.

THE origin of the zeolites and associated minerals in vesicles in lavas is a subject on which there is considerable difference of opinion. In general there are two main views: that they are due to the action of percolating surface waters upon the original minerals of the rock; or are associated with some stage of the cooling processes, as an after-effect of volcanism.

Some years ago the writer made a study of the amygdale minerals in the Tertiary basalt lavas of the south of County Antrim, Ireland. So striking were the differences in the mineral assemblage found during later visits to the east of Antrim that it was felt that important evidence on the origin of the minerals might be obtained if their distribution within the lavas was known. Accordingly construction of a zeolite distribution map of the Antrim basalts was commenced.

As the work progressed the scope widened, and in fact the main purpose of the present paper is to trace the distribution of habit shown by the zeolite chabazite in a selected area of basalts. The distribution of the zeolites associated with the chabazite is also considered. In succeeding papers it is hoped to deal with the distribution of the amygdale minerals in the remainder of the Antrim basalts.

*Previous work.*—In the early days of mineralogy Antrim was one of the classic zeolite areas, but since the mid-nineteenth century, very little has been added to the knowledge of the Antrim zeolite localities, although during the early part of the present century a considerable amount of collecting was done by the late F. N. Ashcroft and R. Bell, and three valuable papers appeared in this Magazine<sup>1</sup> as a result. More recently

<sup>1</sup> F. N. A. Fleischmann, On the occurrence of gyrolite in County Antrim. *Min. Mag.*, 1910, vol. 15, pp. 288–293.

G. F. H. Smith, F. N. Ashcroft, and G. T. Prior, Chabazite and associated minerals from County Antrim. *Ibid.*, 1916, vol. 17, pp. 274–304.

F. N. Ashcroft, The natrolite occurrence near Kinbane (White Head), County Antrim. *Ibid.*, 1916, vol. 17, pp. 305–308.

M. H. Hey<sup>1</sup> used some Antrim material in his studies on the zeolites. As far as our knowledge of the zeolite distribution in Antrim goes, however, the fact remains that little advance has been made since J. E. Portlock published his Report on the geology of Londonderry in 1843. Apart from a very brief mention in the Geological Survey Memoir on the area nothing whatever appears to have been written on the zeolites of the Garron plateau, with which the present work is concerned.

## 2. GEOLOGICAL ENVIRONMENT.

Tertiary plateau-lavas, dominantly basaltic, cover an area of some 1500 square miles in the north-east of Ireland (fig. 1), resting for the

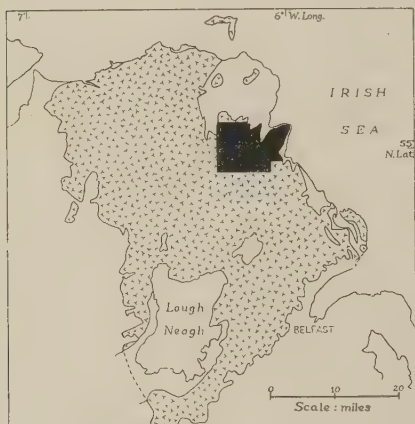


FIG. 1. Tertiary lavas of north-east Ireland (stippled); position of Garron plateau area (black).

most part upon sedimentary rocks of Mesozoic age. Although actually neither confined to the County of Antrim nor composed entirely of basalt the lava series is conveniently designated the Antrim basalts. The greatest known thickness of the lava pile is about two thousand feet, but the lavas have been deeply eroded and over most of the area the thickness preserved for study is much less than this. The constituent lava flows attain a maximum thickness of upwards of one hundred feet, but the average

is of the order of about twenty feet. Evidence of eruption under sub-aerial conditions is afforded by the red lateritic weathering of the tops of many of the flows, by the absence of pillow-lavas, and by the existence in the lava succession of occasional beds bearing plant remains.

In general, each lava flow possesses an amygdaloidal top and a thin amygdaloidal base, the latter commonly with pipe amygdales. Amygdales are often also present in the more massive interior of the flows. The cavities vary considerably in size, but are commonly an inch or more in diameter. It is the writer's experience that empty cavities are

<sup>1</sup> M. H. Hey, Studies on the zeolites. *Min. Mag.* 1932, vol. 23, pp. 51, 243; 1933, vol. 23, pp. 305, 421; 1934, vol. 23, pp. 483, 556; 1935, vol. 24, p. 99; 1936, vol. 24, p. 227.

exceptional; they almost always contain minerals of the zeolite group or their associates, particularly calcite.

*Geology of the Garron plateau area.*—The present work deals with the amygdale minerals in seventy square miles of lavas in the east of County Antrim, comprising the Garron plateau proper and adjoining regions to the west. This district was selected for detailed study for geological and geographical reasons.

The lavas within this area approach horizontality, although there is a slight prevailing dip towards the south. There are few faults. The geological structure is admirably reflected in the topography, in the flat plateau of Garron with average elevation of 1000 feet and bounded on the north-west, north, and east by steep, often precipitous escarpments, and in the long, flat-topped, wedge-shaped ridge of Lurigethan. Towards the north-west the land is rather higher, a consequence of the regional dip, rising to 1800 feet at Trostan.

Exposures are good in the escarpments, up to 1000 feet in thickness of lavas being available for study, but on the flat plateaux the rocks are concealed to a large extent by bog, and exposures of amygdaloidal basalt are largely confined to the water-courses.

Practically all the lavas in the area under consideration are olivine-basalts of rather uniform character, associated with subordinate picrite-basalts and olivine-poor basalts and mugearites. The lava succession has long been separated into two stratigraphical divisions, referred to as the Lower and Upper Basalts respectively, and separated by a thick zone of weathering, the lateritic iron ores and bauxites which were mined extensively in the past. Most workers are agreed that this interbasaltic zone of weathering represents a very considerable time-interval and lull in the eruption of basalts; the figure of one million years has recently been quoted by V. A. Eyles.<sup>1</sup>

Throughout the Garron area the Lower Basalts retain a uniform thickness of 450–500 feet. Individual flows may cover a wide extent; for example, a distinctive flow of basalt with large felspar phenocrysts rather more than midway up in the Lower Basalts may be traced laterally over a distance of at least eight miles. This wide lateral spread of individual flows and resulting comparative uniformity in the lava succession over a wide area deserves emphasis, as also does the uniformity in the composition of the lavas themselves.

<sup>1</sup> V. A. Eyles, Abstr. Proc. Geol. Soc. London, 1950, no. 1465, pp. 132–133.

## 3. ZEOLITES IN THE GARRON PLATEAU AREA.

The following are the zeolites found in the lavas of the Garron plateau area, arranged in order of relative abundance:

*Chabazite*.—Chabazite is ubiquitous. It occurs as distinct crystals ranging up to half an inch or so in size, and unlike the other zeolites never forms radiating aggregates. In optical character it is sometimes uniaxial, but more commonly biaxial with small 2V. The sign is negative when the mean refringence exceeds about 1.488, and is positive when lower. The birefringence is small, usually of the order of 0.003 or less. The mean refringence ranges from 1.483 to 1.495, and it is unrelated to the habit of the crystals. Some crystals are zoned, but the zoning has not been studied in detail.

*Thomsonite*.—Thomsonite is usually closely associated with chabazite, but is rather less common. It characteristically forms compact aggregates of small acicular or bladed crystals, often hemispherical in form and with radiating structure, and commonly as a cavity-lining to the larger cavities.  $\beta$  varies from 1.515 to 1.530. The more or less hemispherical aggregates often show concentric zoning, with lower refringence near the cavity-wall.

The variety 'sphaerostilbite' is less common, forming a distinct second generation, as described from Golden, Colorado, by H. B. Patton,<sup>1</sup> but not differing appreciably in optical properties from the more normal thomsonite with which it is associated, despite the different age and appearance. It typically occurs as non-compact aggregates of very delicate crystals.

*Levyne*.—Levyne is very widespread, nearly always as thin tabular crystals with (0001) prominent, and invariably penetrant-twinned. Sheaf-like aggregates are found occasionally. In optical character it is uniaxial negative, with  $\omega$  normally in the range 1.499 to 1.505. The birefringence is about 0.005.

A very interesting and characteristic feature, shown by at least half of all the crystals in the area is the alteration to a fibrous mineral with satiny lustre, the fibres lying normal to (0001) of the levyne. This mineral is believed to be uniaxial with negative sign and refractive indices  $\epsilon$  1.491–2,  $\omega$  1.495. W. Cross and W. F. Hillebrand<sup>2</sup> recorded a fibrous mineral from Golden, Colorado, closely associated with levyne; how closely is not stated, and there is no mention of it occurring as an

<sup>1</sup> H. B. Patton, Bull. Geol. Soc. Amer., 1900, vol. 11, pp. 461–474.

<sup>2</sup> W. Cross and W. F. Hillebrand, Bull. U.S. Geol. Surv., 1885, no. 20, vol. 3, pp. 231–257.

overgrowth. Their analysis of the fibrous mineral proved to be almost identical with that of levyne from the same locality. They concluded, 'it cannot be referred to any definite species from present data'.

*Analcime*.—Analcime is quite abundant in the area, mostly as single crystals which may exceed an inch in diameter, but occasionally in the form of radiating aggregates. The crystals are always of simple habit, (211) being the only form observed. The mineral is never isotropic, and possesses a birefringence of up to about 0.003. The mean refringence is near 1.487.

*Natrolite, mesolite, and scolecite*.—These three minerals very commonly occur together as tufts of delicate hair-like crystals. They have crystallized in the order listed, mesolite continuing the growth of natrolite at the extremity of the crystals, and scolecite forming a thin overgrowth on the mesolite. Mesolite is almost isotropic, with mean refringence of about 1.504. Scolecite has an appreciable birefringence, with  $\alpha$  1.507,  $\beta$  1.519, and an extinction of about  $16^\circ$ . In addition to the delicate tufts natrolite is not uncommonly found in the form of compact aggregates of outer crystals. In the Garron area natrolite is inconspicuous in amount when compared with analcime: on the other hand, in certain other parts of Antrim the converse is true.

*Other minerals*.—Phillipsite is found at a considerable number of localities, but is seldom abundant or conspicuous. It forms both single crystals and radiating aggregates. Stilbite and heulandite have also been met with in the course of the work, but neither is common. In one or two places the latter has been found as a granular layer on chabazite and calcite. Of the non-zeolites, calcite is very common throughout the area. Aragonite is rare, and quartz has not been observed. Apophyllite has been found at one locality.

*Order of crystallization*.—Levyne is believed to be the earliest zeolite. Certainly whenever it is associated with chabazite or thomsonite in the same cavity it precedes them, but its relationship to phillipsite, which definitely precedes chabazite, is uncertain. Chabazite and thomsonite are very closely associated, sometimes the one being the earlier, sometimes the other, and in all probability their periods of crystallization overlapped. The habit of thomsonite which has been called 'sphaerostilbite' constitutes a distinct generation appreciably later than chabazite. Stilbite and heulandite, when they occur, are later than chabazite. An important feature, which will be referred to again when the origin of the minerals is considered, is that analcime crystallized later than the levyne-thomsonite-chabazite assemblage. At a few localities analcime

is followed by a sugar-grained aggregate of chabazite of low (i.e., simple) habit, representing a distinct generation of chabazite in the same way that the variety 'sphaerostilbite' represents a distinct generation of thomsonite. The latest zeolites in the sequence appear to be natrolite, mesolite, and scolecite. Of the non-zeolites, calcite may either precede or follow the zeolites. Apophyllite, in the single locality at which it has been found, is later than thomsonite and analcime.

#### 4. CHABAZITE CRYSTAL HABITS.

Chabazite is a trigonal mineral with the axial ratio  $a : c = 1 : 1.086$ . Twinning may take place on two laws, namely contact twinning on  $(10\bar{1}1)$  and interpenetration twinning on  $(0001)$ . The crystals show a considerable range of habit, from the simple, untwinned unit rhombohedron to the complex-twinned varieties, phacolite and herschelites. Different habits may be recognized, based on the relative development of some half-dozen different forms and the incidence and nature of the twinning. These cannot be regarded as completely unrelated habit elements as, to take an example, when  $s(02\bar{2}1)$  is present it is usually accompanied by  $e(01\bar{1}2)$ , vicinal faces on  $r(10\bar{1}1)$ , and interpenetration twinning on  $(0001)$ . It is this interrelation of elements that has enabled a habit series to be recognized; as a result of the examination of many thousands of crystals it appears certain that there is a continuous gradation between two extremes, and that all, or practically all, chabazite crystals may be arranged in this simple habit series.

The simplest crystals show the unit rhombohedron,  $r(10\bar{1}1)$ , alone or commonly with the faces curved and stepped, and twinning on  $r$  is very common. Tracing the habit series in the direction of increasing complexity, interpenetration twinning on  $(0001)$  first appears, the twin individuals being of unequal size at first, and the curvature and stepping of the faces and the twinning on  $r$  are no longer found. The next change is the partial replacement of  $r$  by vicinal faces, which appear as striations parallel to the terminal edges of the unit rhombohedron, and faces of other forms then appear,  $s(02\bar{2}1)$ ,  $e(01\bar{1}2)$ , and sometimes  $a(11\bar{2}0)$ . Usually  $s$  precedes  $e$ . The relative size of the twin individuals also becomes more uniform. Continuing, the relative sizes of  $s$  and  $e$ , both normally present at this stage, gradually increase and  $a$ , if present, becomes very prominent,  $r$  at the same time decreasing in size and importance. The vicinal faces in the zone  $re$  become better developed and  $t(11\bar{2}3)$  usually appears at this stage and may become prominent.

Soon after the appearance of  $t$  the well-defined shape of the crystals

becomes lost and the faces of  $r$ ,  $e$ , and  $t$  become broken up into innumerable minute facets in oscillatory combination:  $a$  (when present) and to a lesser extent  $s$  resist this tendency to break up and persist as discrete faces. The crystals assume the overall shape of a flat hexagonal pyramid, the 'pyramid' faces being composed either of  $t$  or of  $r$  and  $e$  in oscillatory combination. Crystals of this habit have long been termed 'phacolite' in reference to their lenticular shape, and although the term has been used at times in a wider sense it seems desirable to limit the term, as here, to the complex-twinned habit with  $r$  and  $e$  represented by minute facets in oscillatory combination rather than as discrete faces or, alternatively, to crystals in which  $t$  is the predominant form.

The final habit of the series would appear to be that which is marked by the appearance of  $c(0001)$  on crystals of phacolite habit; and the rare variety 'herschelite', with  $c$  and  $s$  as practically the only forms present, is probably the most extreme habit. In the Antrim basalts chabazite is the most common and characteristic zeolite, and crystals of all the above habits, with the exception of typical herschelite, are abundantly represented.

*Chabazite habit notation.*—For the purpose of recording chabazite habit data a simple descriptive notation has been developed in which the crystals are classified into six habits, defined as follows:

*Habit 1.*— $r$  alone. No interpenetration twinning. Commonly present: simple twinning on  $r$  and stepping and curvature of the faces.

*Habit 2.*— $r$  alone, without vicinal faces. Interpenetrant-twinned.

*Habit 3.*— $r$  dominant but normally striated by vicinal faces, and other forms present ( $s$ ,  $e$ , and sometimes  $a$ ). Interpenetrant-twinned.

*Habit 4.*— $r$  subordinate, and other forms ( $e$ ,  $s$ ,  $a$ ,  $t$ , and vicinal faces in the zone  $re$ ) dominant. Interpenetrant-twinned. Forms represented by discrete faces.

*Habit 5* (phacolite habit).—Crystals with overall lenticular shape, complex interpenetration twinning with  $r$ ,  $e$ , and  $t$  as minute facets in oscillatory combination, or alternatively crystals with  $t$  prominent.  $a$  may or may not be present;  $s$  always present.

*Habit 6* (herschelite habit).—Characterized by the presence of  $c(0001)$ . Not represented in the Garron area.

The above habits are illustrated by the drawings of fig. 2. Some drawings are idealized; particularly is this true of habit 4 in which marked irregularities of growth are usually present. It has been decided

to refer to the more complex crystals as possessing 'higher' habits, and the more simple crystals as 'lower' habits.

*Chabazite habit study in the field.*—It has been found that, in general,

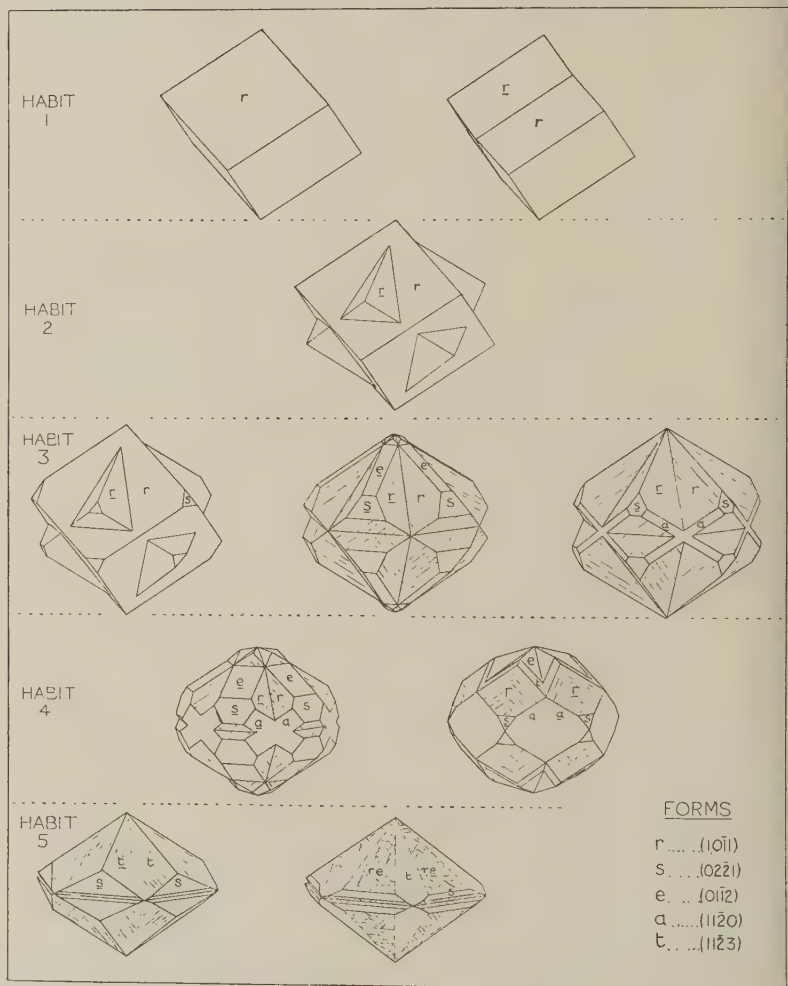


FIG. 2. Chabazite habits, illustrating the proposed habit notation.

the chabazite crystals in any one cavity all possess approximately the same habit, and further that, at a given locality, all the crystals will possess very nearly the same habit. The extent of such variation as does

occur is illustrated by the diagrams of fig. 3, which embody the results of a study of the habit variation at four localities in County Antrim.

To illustrate the procedure, a quarry on the Collin Mountain in the south of Antrim may be considered (*a*, fig. 3). A large number of chabazite-bearing cavities were collected from the red amygdaloidal top of a flow and from the basal part and interior of the overlying flow. It was found that in no cavity did habit 1 occur alone; in 85 % 1 and 2 were present together, 1 being predominant in half of them, 2 in the other half; and in only 15 % of the cavities did habit 2 appear alone. In no case was any form other than  $r(10\bar{1}1)$  seen. When habits 4 and 5 appear they are usually associated with a rather greater range of habit (*c*, fig. 3).

Having established that crystals at a given locality show a limited variation in habit, the possibility arises of mapping the lateral and vertical distribution of habit in the lavas. Assuming the crystals are amenable for habit study such a three-dimensional picture of the habit distribution can be drawn only if the mineral is sufficiently widespread and if there are abundant exposures both laterally and vertically through an appreciable thickness of lavas. Further, the exact position of any locality in the lava series must be determinable, involving an accurate knowledge of the stratigraphy and structure of the lavas. In Antrim such geological conditions are perhaps realized only in the Garron plateau area, where chabazite is fortunately very abundant.

##### 5. CHABAZITE HABIT DISTRIBUTION IN THE GARRON PLATEAU AREA.

It has been possible to recognize five chabazite habit assemblages for purposes of mapping, and these have been utilized on the map of the Garron plateau area (fig. 4). The 227 localities on which the habit zones on the map are based have been plotted to illustrate the degree of control achieved. The habit distribution is found to be extremely regular.

Habit 5, usually associated with lower habits (that is, habits 4, 3, and sometimes 2), occurs only towards the base of the lavas and has been found at six places: in the Bush valley, in Glen Ballyemon, in small patches near the Altmore Burn and Evish, and in larger expanses in the

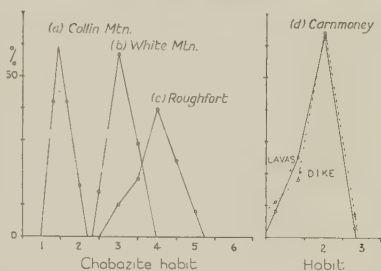


FIG. 3. Variation of chabazite habit at four localities in County Antrim.



vicinity of Garron Point and near Carnlough. At several of these places, notably near Altmore Burn, habit 5 is found in the middle of the Lower Basalts rather than at the base, an observation that assumes significance when the origin of the minerals is considered.

Habit 4, also usually associated with lower habits, is distributed in a zone enclosing the previous one, and naturally has rather wider extent; in fact in only a few places is habit 4 missing from the lower part of the lava series. The zone characterized by the presence of habit 4 is almost entirely confined to the lower 350 feet of the Lower Basalts, but probably reaches the interbasaltic horizon near Garron Point, and almost reaches it in the Cranny Water.

The third chabazite habit assemblage is characterized by the predominance of habit 3, usually associated with habit 2 in smaller amount, but without more than a very small amount of habit 4. Habit 3 falls into a continuous zone, probably nowhere exceeding a height of 550 feet in the lavas, but crossing the interbasaltic horizon north-west of Trostan, at Tuftarney Hill, near the Inver River, and in small areas near Garron Point and Cranny Water.

The next habit assemblage is characterized by the predominance of habit 2, often in the presence of minor amounts of habits 1 or 3. This habit again falls into a continuous zone and is usually located near the interbasaltic horizon, although locally entirely above or below it. The zone varies from fifty to several hundred feet in thickness.

The two remaining habit assemblages both contain an abundance of habit 1, either in association with habit 2, the two being in comparable amounts, or alone. Habit 1 occurs alone, or in association with a minor amount of 2 in a thick zone, the top of which is not exposed, forming the top of the Garron plateau proper. The isohabit surface forming the base of this zone has a marked regional easterly dip with respect to the lavas, being absent from the top of Trostan (900 feet up in the lavas) and the hills south of Cargan, coming in at a height of some 800 feet at Carn Neill, and reaching to a height of only 400 feet in the lavas, that is, below the interbasaltic horizon, north of Carnlough. The assemblage characterized by the association of habits 1 and 2 in comparable amount forms a continuous zone of variable and often considerable thickness between those of habits 3 and 1 alone respectively, and forms the tops of the principal hills in the west of the Garron area.

The chabazite habit data which have been collected are treated statistically in the diagrams of fig. 6, where the distribution of habits at nine different levels in the lavas has been plotted. It can be seen that

habits 4 and 5 are practically confined to the lower 450 feet of lavas, and that habit 3 occurs mostly in this range, whereas habit 1 is mostly confined to the lavas above 450 feet in the succession. Habit 2 is slightly more abundant in the higher levels, but does not exhibit any very marked preferential distribution, a consequence of its presence in variable and often small amount in all, or nearly all of the habit assemblages recognized for mapping purposes.

Summing up, the habit shown by the chabazite varies but little at individual localities, and the habit assemblages which are found in the lavas fall into a number of well-defined zones. These zones have a rough stratigraphical distribution, but they are clearly superimposed upon the lava pile just as zones of regional or contact metamorphism characterized by certain mineral assemblages, or zones in a mineralized area such as Cornwall or Derbyshire and characterized by a certain gangue or ore mineral, are superimposed upon the country-rock.

*Significance of habit distribution.*—The genetic significance of this habit distribution is clear: the chabazite is entirely later than the 1000-feet or so of lavas in the Garron area. Further valuable evidence of genetic significance is derived from a study of the several faults which cut the lavas of the Garron plateau. Of these the Cushenilt and Galboly faults are magnificently exposed in the cliffs west of Garron Point, and each has a throw of rather more than 200 feet, which is readily demonstrated in the field. After careful study of the chabazite habits on either side of these faults and also of the smaller fault in the Inver Burn it has been established that the chabazite habit zones are faulted to the same extent as the lavas themselves. There is evidence, in the occurrence of chabazite (of lower habit than in the adjacent lavas) in the fault fractures, that the formation of chabazite did not entirely antedate the faulting, but the fact remains that the bulk of the chabazite must have preceded the faulting.

The relationship between chabazite habit zones and faults is best seen in the section, fig. 5 (upper part). This has been constructed from the data from 88 zeolite localities distributed along the northern escarpment of the Garron plateau proper, from the Inver River near its intersection with the interbasaltic horizon through Evish to Garron Point. Although clearly not on a vertical plane the error resulting from the localities lying on a sloping surface is probably negligible.

The conclusions reached after a study of the distribution of chabazite habits is that the bulk of the chabazite crystallized after the exposed lava pile had been erupted but before it was faulted. The possibility that

the chabazite crystallized during the cooling of individual flows immediately after their eruption may be excluded at once as incapable of accounting for the observed habit distribution, and that the chabazite originated as a result of weathering by present-day percolating waters of meteoric origin may equally definitely be excluded, particularly in view of the faulting of the habit zones.

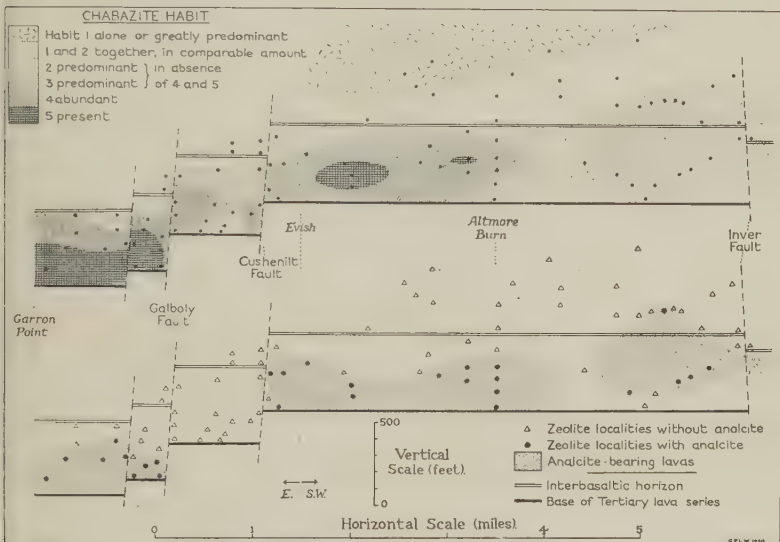


FIG. 5. Sections along the north escarpment of the Garron plateau showing distribution of chabazite habits (upper section) and analcime (lower section). Based on 88 zeolite localities.

Considering the reason for the observed habit distribution, variation of habit in crystalline substances can be related to the operation of a variety of factors, of which the temperature, rate of crystallization, and the presence and nature of impurities are perhaps the most important. The writer considers that the temperature of formation is most likely to have been the controlling factor in the development of the different habits in the case of the Garron chabazite, on account of the extreme regularity of the habit distribution.

It is tempting to regard chabazite of habit 5 as having been formed at the highest temperature, with the lower habits crystallizing at progressively lower temperatures. Supporting evidence is provided by the fact that, whenever two generations of chabazite can be distinguished, the later is always of lower habit than the earlier. The use of this as evidence

of temperature control involves, of course, the not unreasonable assumption that the later chabazite formed at a lower temperature than the earlier. Temperature control of the habit is capable of accounting for the common association of habit 5 with lower habits.

#### 6. ZEOLITE DISTRIBUTION IN THE GARRON PLATEAU AREA.

Chabazite is almost ubiquitous in the Garron plateau area and is evenly distributed throughout the lava series. Certain of the other

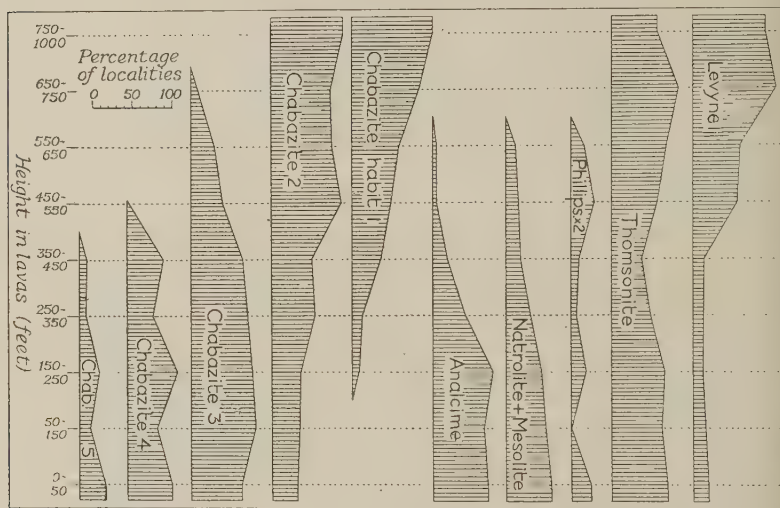


FIG. 6. Distribution of chabazite habits and zeolites at different levels in the lavas of the Garron plateau area. Graphs based on data from 243 zeolite localities.

zeolites, however, have restricted distribution in the lavas. To study the distribution of these minerals the records of the zeolites present at each of the 250 zeolite localities in the area have been analysed, and the percentage of localities in each of the nine arbitrary levels in the lava series at which the particular zeolite has been recorded is plotted on the diagram, fig. 6. It can be seen that analcime, natrolite, and mesolite (with scolecite) are mainly confined to the lower few hundred feet of lavas; levyne, on the other hand, is mainly confined to the upper levels. Thomsonite and phillipsite have also been plotted, but show little sign of any preferential distribution.

The distribution of analcime is particularly regular and interesting. The mineral has been recorded at 75 of the 120 localities in the lower

350 feet of lavas, and in only seven of the 120 or so localities above 350 feet. In the lower part of the basalts analcime is sufficiently abundant to enable a distribution map to be drawn (fig. 7). The areas in which analcime occurs have been stippled, and as in the chabazite habit distribution map, the position of the localities upon which the distribution of the stippling has been based are given to illustrate the degree of control achieved, which varies in different parts of the map. The lower

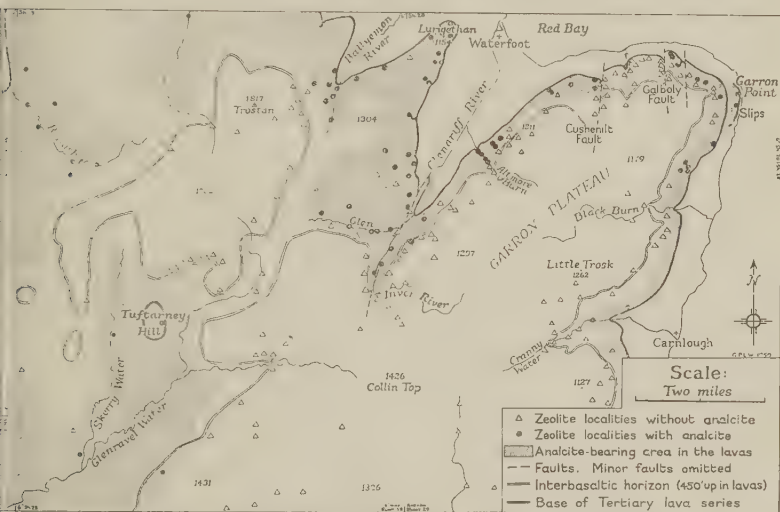


FIG. 7. Distribution of analcime in the lavas of the Garron plateau. Based on 228 zeolite localities.

section of fig. 5 can be used in conjunction with the map. The main points to notice are the confinement of analcime to an almost continuous zone in the lower part of the lava succession, and the fact that this zone is not a stratigraphical horizon such as might be expected were analcime confined to a certain type of lava. The analcime zone is clearly superimposed upon the lava pile, rising above the interbasaltic horizon at three places (south of Glenravel Water, at Tuftarny Hill, and north of the Inver Burn), being probably absent from the lavas at one point near the Altmore Burn, very thin in the vicinity of Carnlough, and definitely absent between the Cushenilt and Galboly faults.

The disappearance of the analcime zone between the faults is particularly significant, and can hardly be due to chance. It will be remembered that, although the bulk of the chabazite antedated the faulting, some

chabazite has been deposited in fissures associated with the faults. As analcime is known to have succeeded chabazite it would be expected to have post-dated the faulting, and certainly the analcime zone shows no indication of having been faulted. However, analcime is not found in the fault fractures or in the immediate vicinity of the faults. It seems as though the conditions which obtained in the lavas near to the faults were inimical to the formation of analcime, and the suggested explanation is that the fault fractures provided a path for the escape of hot waters from the lower parts of the lavas, thus lowering their temperature in the vicinity of the faults to below that at which analcime was able to form.

It is not certain what were the factors controlling the distribution of levyne, which varies in amount inversely with the analcime, natrolite, mesolite, and scolecite. It seems very likely that the small content of levyne in the lower part of the lavas is due to its destruction or concealment by the later minerals, and in particular by analcime.

#### 7. ORIGIN OF THE ZEOLITES.

Most workers are agreed that the zeolites in amygdaloidal rocks have derived their material from these rocks, from the feldspars, or glass when present. Glass is absent from the lavas of the Garron plateau area, and the feldspar is indicated as the most likely source of the materials which compose the zeolites. The chloritic minerals which are seen in thin sections of the lavas represent the hydration products of the other pyrogenetic minerals of the basalt. There is sometimes evidence of the enlargement of zeolite-bearing amygdales by corrosion of the adjacent basalt, but it is likely that the bulk of the zeolites in the cavities crystallized from material migrating from the surrounding rock.

It is generally agreed that the zeolites in lavas were formed at temperatures normally above  $100^{\circ}$  C. It would seem, then, that a source of hot water is required to account for the production of the zeolites in the Garron lavas. The most obvious source of heat is that supplied by the lavas themselves. However, the evidence of the distribution of chabazite habits and analcime shows that the zeolites, or at any rate the bulk of them, were formed not within individual flows as they cooled immediately after eruption but after a considerable thickness (at least 1000 feet) of lavas had accumulated.

On the eruption of a lava flow cooling takes place both upwards into the atmosphere and downwards into the subjacent rocks. There is therefore the possibility of the progressive accumulation of heat in the lava pile by a succession of small increments from each flow erupted.

However, the deep lateritic weathering of the tops of many of the flows shows that a considerable time interval often elapsed between the eruption of successive flows, and the interbasaltic horizon in particular represents a lull in the eruption of flows of considerable duration. In the circumstances it is probable that no appreciable accumulation of heat in the lavas could be expected by this means.

An alternative is to invoke the agency of heated waters rising from depth. This would be more plausible were there abundant faults in the Garron area to provide a passage for these waters; as it is, there is very little fracturing. The lavas of the Garron area are cut by a number of dikes with north-west trend, part of the great British Tertiary dike swarm, and the field evidence suggests that most of these dikes are later than most of the exposed lavas. It is possible that the dikes added a considerable amount of heat to the lava pile when intruded into it, and they may have caused an appreciable rise in the geoisotherms, already higher than in non-volcanic regions. However, the density of the swarm is not very great in the Garron area, there being probably no more than two or three dikes to the mile across the strike, and it seems unlikely that the heat from the dikes could have been adequate to give rise to such widespread development of zeolites. It is perhaps possible that rhyolite sills intruded into the Mesozoic or earlier sedimentary rocks below the lavas may have contributed, but no sills are present in the several feet of sedimentary rocks exposed in the district.

A further possibility is that the source of heat lies in the reactions producing the zeolites. The hydration of the feldspars to zeolites is probably an exothermic reaction; so is the hydration of the olivines and pyroxenes. It is suggested that the heating up of the lava pile by dikes and other intrusions, by regional uprise of deep-seated waters, or by conduction of heat downwards from later erupted lavas initiated the reactions in the pyrogenetic minerals of the basalts and that, once started, sufficient heat was generated by these reactions to maintain them and to heat up the lavas further, the reactions perhaps coming to an end only when all the available water was used up.

This conception of self-generation of heat in the lava pile appears capable of accounting for certain features in the distribution of chabazite habits and zeolites that would be difficult to explain otherwise. If heat were generated throughout the lava pile there would be loss of heat towards both by conduction and by migration of heated waters. As it is unlikely that a state of thermal equilibrium would be attained, which would involve the raising of the temperature in all the underlying rocks

to more than that in the lavas, there would probably always be a certain amount of conduction of heat downwards from the lavas as well as upwards. Under such conditions the maximum temperature in the lava pile would be attained in the lower part but not quite at the base, and there would be a slight decrease in temperature from there to the base of the lavas. Upwards from the maximum there would be at first a slow falling-off in temperature becoming more rapid upwards. There is evidence from the distribution of chabazite habits and analcime that assuming temperature to be the controlling factor, the maximum temperature was reached somewhere between the base of the lavas and a height of about 300 feet.

#### 8. IMPLICATIONS OF THE WORK.

Mineralogists have always recognized that crystals of the same mineral often exhibit a considerable range of habit, yet the rapid progress in mineralogy of the last century has not advanced appreciably the knowledge of the significance of these various habits. In the present work an attempt is made to approach the question by studying the spatial distribution of habit shown by a mineral in a selected area of lavas, and the resulting chabazite habit map (fig. 5) is perhaps the first of its kind yet produced. The distribution is surprisingly regular, so much so as to suggest that the isohabits bounding the habit zones were parallel to geoisotherms and that the habit was controlled primarily by the temperature of formation of the mineral.

In addition to the crystallographic significance of the study of habit distribution the work has an important bearing on the age of the zeolitization, and enables a relative chronological sequence of events to be recognized. In the Garron area 1000 feet of lavas are exposed, comprising perhaps forty to fifty distinct flows, the accumulation of which must have taken at least several millions of years. The chabazite zones are superimposed upon the lavas, and the chabazite is thus indicated as having crystallized after the eruption of this lava pile. Towards the end of the period of chabazite formation the lavas were faulted, and at some subsequent period analcime and the fibrous zeolites natrolite, mesolite and scolecite were formed. Finally, erosion carved out the present-day topography, the valleys being excavated not only through the lavas but also through the zeolite zones.

Unfortunately the time scale arrived at is purely relative, and it is not certain at what part of the Tertiary period the zeolitization and faulting took place. However, it seems to be significant that the newer Tertiary

basalts of the Columbia plateau and the Hawaiian Islands contain zeolites only exceptionally. It may be that the process of zeolitization is a very slow one, and that these lavas are not of sufficient antiquity to have allowed the process to have taken place on a large scale. If, as is suggested here, the hydration of the pyrogenetic minerals of the basalts to zeolites and chlorites is capable of generating sufficient heat to maintain the reactions once initiated, and to account thus for the zeolitization, it is probable that the rise in temperature of the lavas to a sufficiently high level would require the presence of a considerable cover of lavas or other rocks, and that a very long period of time would be required before the reactions could get under way. The hot springs in certain volcanic areas, excluding those associated with volcanoes of central type, may represent the surface manifestation of present-day zeolitization below the surface.

In the Garron area the faulting took place more or less in the middle of the period of zeolitization. This has been demonstrated only in the case of several faults and the work requires to be extended to a wider area before any far-reaching conclusions can be drawn, but it may be that the Garron faults were formed at the time of the break-up of the great North Atlantic lava plateaux. One possible application of the work is the dating of faults and the grouping of faults into time groups, and, taking into account the relationship between the inclination of lava flows which have been tilted subsequent to their eruption and the zeolitization, it may be possible to gain an insight into the post-volcanic history of the lava plateaux. Another application of the methods described in this paper is the location, under favourable circumstances, of faults the exact position of which is uncertain. A third possibility is that continued study of chabazite habit distribution, coupled with experimental work, might provide a means of enabling estimates to be made of the original thickness of the lava pile in selected areas.

*Acknowledgements.*—I am indebted to Professor J. K. Charlesworth and the Research Fund Committee of the Queen's University of Belfast for several grants towards travelling expenses; to Professor W. Q. Kennedy, in whose Department the laboratory work was done, for reading the manuscript; and to Dr. W. Campbell Smith and Dr. M. H. Hey for reading the manuscript and for facilities afforded for examination of the zeolite collection of the British Museum.

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*On crossite from Anglesey.*

By NORMAN HOLGATE, M.Sc., Ph.D., F.G.S.

Department of Geology, University of Glasgow.

[Read November 1, 1951.]

*Introduction.*

THE occurrence of a member of the glaucophane group of minerals in Anglesey has been known since 1888, in which year Blake<sup>1</sup> reported the presence of a blue amphibole in a schist exposed in the neighbourhood of the Anglesey Column, west of the Menai Bridge. In the course of his paper Blake made general remarks on the petrography of the schist and on its field relations. He dwelt at some length on the question of the identity of the blue amphibole which is its most notable constituent, describing its columnar habit and pleochroism, and recording an extinction angle of  $15^\circ$  with the axis of elongation (*c*). By comparison with a description of glaucophane due to Rosenbusch, Blake concluded that the blue amphibole from the Anglesey rock is a normal glaucophane, although its extinction angle as recorded by him is considerably in excess of the  $4^\circ$  to  $6^\circ$  mentioned by Rosenbusch. The associated minerals he identified as epidote, rutile (?), quartz, a feldspar (?), and some calcite (the marks of interrogation are his own).

The chemistry of the glaucophane-schist was subsequently studied by Washington,<sup>2</sup> who gave a short description of the rock analysed. He stated that the glaucophane-schists 'are seen to be composed of a rather pale, prismatic glaucophane, with the normal pleochroism, rather less [amounts of] pale epidote, and small amounts of magnetite, quartz, mica and titanite'. He also noted the absence of calcite. He regarded the rock as having been derived from a diorite or possibly from a gabbro but was uncertain whether the development of glaucophane-schist in Anglesey was due to regional or to thermal metamorphism.

Greenly, in his classical work,<sup>3</sup> added little to Blake's description of the schist or of its blue amphibole, which latter he, like Blake, considered to belong to glaucophane proper. Greenly's main contribution lay in his investigation of the field relations of the glaucophane-schist

<sup>1</sup> J. F. Blake, *Geol. Mag.*, 1888, dec. 3, vol. 5, pp. 125-127.

<sup>2</sup> H. S. Washington, *Amer. Journ. Sci.*, 1901, ser. 4, vol. 11, pp. 42, 43.

<sup>3</sup> E. Greenly, *The geology of Anglesey*. *Mem. Geol. Surv. Great Britain*, 1910, pp. 115-118. [M.A. 1-328.]

which he clearly distinguished as being products of regional metamorphism.

The identification of the blue amphibole from the schists of the Monument Hill does not appear to have been questioned, and no further communications on the subject have appeared to date. A chemical analysis of the blue amphibole was obviously required, but, since the fineness of grain of the schists which contain it render difficult a reasonably pure separation of the mineral, it is not surprising that no such analysis has previously been recorded. Recently, however, the methods of separating minerals from crushed rocks have been considerably improved, notably by the introduction of the use of the centrifuge in association with heavy liquids. Using this technique, it has been found possible to separate the blue amphibole in a very nearly pure state.

*Petrography of the 'glaucophane'-schist.*

The material investigated was collected from the foot of the crag on the south-eastern face of the Monument Hill, on the north side of the Holyhead road, near the village of Llanfairpwllgwyngyll, and some one and a half miles west of the Menai Bridge.

In the hand-specimen it is a blue-grey rock showing a nemablastic schistosity, with very subsidiary sericitic material intersecting the texture in thin and impersistent films. In thin slice it is seen to correspond closely to the material described by Blake and by Greenly, showing a strongly developed alinement of slender prisms of the blue amphibole and of prismatic to granular epidote, with occasional granular sphene. Also conforming to the schistose fabric are occasional thin layers of quartz and albite mosaic, the latter mineral occasionally twinned on the albite law and sometimes building small sub-porphyritic knots or augen about which the nemablastic fabric is disturbed. A pale green chlorite which is present in small quantities is not recognizably pseudomorphous after any mineral, but is intersected by sharply bounded prisms of the blue amphibole which may have grown in part at the expense of the chlorite. Rutile and calcite referred to by Blake (loc. cit.) have not been observed in the thin slices, nor has been the sericitic material observed in the hand-specimen. Spongy haematite, showing red in thin plates, occurs sporadically in the rock.

*Physical properties of the blue amphibole.*

The habit of the blue amphibole corresponds to the description given by Blake (loc. cit.), the forms (110) and (010) being clearly defined while the terminations are indefinite. It is moderately pleochroic in thin

section, with  $\alpha$  pale neutral tint,  $\beta$  pale grey-violet, and  $\gamma$  sky-blue, the absorption being  $\alpha < \beta < \gamma$ . The refractive indices for sodium (D) light are  $\alpha$  1.649,  $\beta$  1.656, and  $\gamma$  1.657. The optic axial plane is parallel to (010) with  $\beta = b$ , the maximum measured extinction angle  $\gamma : c$  being  $11^\circ$ . The optic axial 2V is variable about  $17^\circ$ , with  $Bx_a = \alpha$ .

As a powder the mineral is medium grey-blue in colour; its specific gravity (determined by dispersing the powder in a liquid of matching density and confirming the stability of the suspension with the centrifuge) is 3.206 (uncorrected value determined at room temperature about  $15^\circ\text{C}.$ ).

While perhaps 90 % by volume of the blue amphibole content of the 'glaucophane'-schist corresponds to the foregoing description, occasional cores of blue amphibole show stronger coloration with  $\alpha$  pale neutral tint,  $\beta$  grey-green, and  $\gamma$  greenish-blue, a formula which suggests a more riebeckitic amphibole than that which it accompanies and which is the subject of this study.

The sample prepared for analysis contained not more than 1 % of epidote as its only impurity. The more strongly coloured amphibole detected in the examination of thin sections had apparently differed sufficiently in specific gravity from the more abundant variety to be completely removed during the separation process.

#### *Constitution and affinities of the blue amphibole.*

The chemical analysis of the blue amphibole is tabulated under 1, table I, and is restated in terms of 'atomic' proportions, adjusted to the conventional basis of  $24(\text{O},\text{OH})$  per unit cell, under 1, table II. The analysis confirms the close relationship of the mineral to the alkali-amphiboles of the glaucophane-riebeckite series.

Analysed glaucophanes agree in showing a uniformly *low* value for the atomic ratio of ferric iron to aluminium, while in the same mineral the ratio of ferrous iron to magnesium varies from fractional values up to a maximum of about unity. In the riebeckites, on the other hand, both of these ratios have high values. The blue amphibole of the Anglesey 'glaucophane'-schist occupies an intermediate position in that the appropriate value for both of these ratios is near to unity, the proportion for aluminium substituted for silicon in the amphibole structure being ignored for the purpose of this comparison. The approximately equivalent proportion of ferric iron to aluminium, and of ferrous iron to magnesium is a character shared with analysed crossites from Berkeley, California and elsewhere. Analyses of Berkeley material by Palache<sup>1</sup> (the type

<sup>1</sup> C. Palache, Bull. Dept. Geol. Univ. California, 1894, vol. 1, pp. 181-192 [Min. Mag. 11-35.]

material) and by Kunitz<sup>1</sup> are quoted for comparison under A and B respectively in tables I and II.

TABLE I. Chemical analyses of crossite.

|                                | 1.           | A.           | B.           |
|--------------------------------|--------------|--------------|--------------|
| SiO <sub>2</sub>               | 50.41        | 55.02        | 54.72        |
| TiO <sub>2</sub>               | 1.66         | —            | —            |
| Al <sub>2</sub> O <sub>3</sub> | 7.82         | 4.75         | 8.27         |
| Fe <sub>2</sub> O <sub>3</sub> | 8.73         | 10.91        | 7.03         |
| FeO                            | 10.81        | 9.46         | 11.82        |
| MnO                            | 0.14         | trace        | 0.36         |
| MgO                            | 7.39         | 9.30         | 7.26         |
| CaO                            | 3.99         | 2.38         | 1.58         |
| Na <sub>2</sub> O              | 7.04         | 7.62         | 6.31         |
| K <sub>2</sub> O               | 0.57         | 0.27         | 0.40         |
| H <sub>2</sub> O + 105° C.     | 1.17         | —            | 1.96         |
| H <sub>2</sub> O — 105° C.     | 0.10         | —            | —            |
|                                | <u>99.83</u> | <u>99.71</u> | <u>99.71</u> |
| Sp. gr.                        | 3.206        | 3.126—3.16   | 3.184        |

1. From crossite-epidote-quartz-albite-schist at base of crag south-east of the Anglesey Monument, Llanfairpwllgwyngyll, Anglesey. Analyst, N. Holgate.

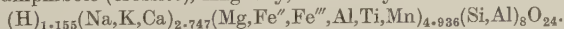
A. From albite-crossite-schist, Berkeley, California. Analyst, W. S. T. Smith (Palache, 1894).

B. From crossite-mica-schist, Berkeley, California. Analyst, W. Kunitz (Kunitz, 1930, p. 244).

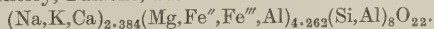
TABLE II. Analyses of crossite, recalculated as atomic proportions.

|       | 1.           | A.           | B.           |
|-------|--------------|--------------|--------------|
| Si    | 7.443        | 7.439        | 7.825        |
| Al    | 1.357        | 0.756        | 1.396        |
| Fe    | 0.185        | —            | —            |
| Fe''' | 0.974        | 1.114        | 0.760        |
| Fe''  | 1.331        | 1.066        | 1.418        |
| Mg    | 1.637        | 1.887        | 1.552        |
| Mn    | 0.009        | —            | 0.043        |
| Ca    | 0.632        | 0.346        | 0.244        |
| Na    | 2.007        | 1.991        | 1.754        |
| K     | 0.108        | 0.047        | 0.073        |
| H     | 1.155        | —            | 1.877        |
|       | <u>1.155</u> | <u>1.155</u> | <u>1.155</u> |

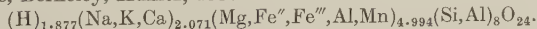
1. Blue amphibole (crossite), Anglesey, new analysis.



A. Crossite, Berkeley, Palache, 1894.



B. Crossite, Berkeley, Kunitz, 1930.



<sup>1</sup> W. Kunitz, Neues Jahrb. Min., Abt. A., 1930, vol. 60, pp. 198, 244. [M.A. 4-100.]

The close similarity of the Anglesey material to these named crossites, on the basis of chemical composition, is well shown by a comparison in detail. While members of the glaucophane-riebeckite series proper do not bear any considerable content of lime, the Anglesey mineral, in agreement with the Berkeley crossites though in greater degree, carries lime in such proportions as to take a significant part in the constitution of the mineral. Unless, therefore, this unusually high lime content is regarded as sufficiently important to constitute a diagnostic character, in which case a new mineral name would be called for, the Anglesey blue amphibole, hitherto regarded as a true glaucophane, should be recognized as an outlying member of the crossite group of the alkali-amphiboles. Its assignment to crossite rather than to glaucophane is in agreement with Sundius's<sup>1</sup> (p. 17) proposal based on the atomic ratio of ferric iron to that part of aluminium occupying positions of six-coordination in the amphibole structure. According to this criterion, Kunitz's crossite (analysis quoted under B, tables 1 and 2) is rather to be referred to glaucophane.

The recalculated analysis of the present material corresponds closely to the general formula for the alkali-amphiboles proposed by Kunitz (loc. cit.) and by Warren.<sup>2</sup> The recalculation shows aluminium proxying for silicon in very nearly the same proportion as does calcium appear in the unit cell. This would support the suggestion due to Winchell<sup>3</sup> that there is present in these minerals a lime-bearing hornblende molecule.

The determination of combined water ('+105° C.') was made by the Penfield tube method, and hence may be in part responsible for the low value of hydroxyl shown in the recalculated analysis. Yet the apparatus used and the procedure followed in the determination might have been expected to achieve a nearly complete expulsion of the combined water of the sample, and its subsequent weighing. An alternative possibility, that the permissible total hydroxyl content is to a considerable degree substituted by halogens, has not been investigated.

A consideration of the physical properties of the Anglesey blue amphibole gives further support to the conclusion reached on chemical grounds. Kunitz (loc. cit.), in his tabulation of the 'glaucophane series', records specific gravities which vary from 3.085 for a glaucophane from the Matterhorn to 3.382 for riebeckite from Pike's Peak, Colorado. The

<sup>1</sup> N. Sundius, *Årsbok Sveriges Geol. Undersök.*, 1946, vol. 40, no. 4. (Avh. Upps. Ser. C., no. 480.) [M.A. 10-70.]

<sup>2</sup> B. E. Warren, *Zeits. Krist.*, 1930, vol. 72, pp. 493-517. [M.A. 4-278.]

<sup>3</sup> A. N. Winchell, *Elements of optical mineralogy*. Pt. II, New York, 1933, p. 259. [M.A. 5-337.]

Anglesey crossite with its specific gravity of 3.206 lies slightly above the value (3.184) determined by Kunitz for a crossite from Berkeley.

The refractive indices of the Anglesey crossite similarly accord well with those recorded for the Berkeley crossites, the values for the former lying between the respective values determined by Palache and by Kunitz. The birefringence of the Anglesey mineral is somewhat low by comparison.

The optical orientation of the crossites appears to be less useful as a diagnostic character, since there is some lack of unanimity on this matter as between described crossites. The Anglesey mineral has its optic axial plane parallel to (010) and the obtuse bisectrix  $\gamma$  near to the crystallographic axis  $c$ . The original crossite described by Palache (loc. cit.) is given as having its optic axial plane also parallel to (010), but as its optic symmetry axis  $\alpha$  near to  $c$ ; the optic sign is not given (Kunitz, loc. cit., p. 198, appears to have misquoted Palache's determinations). According to Louderback,<sup>1</sup> the optic orientation of another crossite from the Berkeley area has its optic axial plane normal to (010) with great dispersion of the optical axial plane about  $b$ . Murgoci,<sup>2</sup> following Brouwer,<sup>3</sup> states that crossite is characterized by having the optic axial plane normal to (010). Sundius (loc. cit., p. 17) states that while those members of the glaucophane-riebeckite series with the ratio  $\text{Fe}^{+++}/\text{Al}$  for positions of octahedral coordination less than unity are characterized by the orientation of the optic axial plane parallel to (010), the members (crossites, riebeckites) for which the ratio exceeds unity, while tending to have the optic axial plane normal to (010), do not invariably show this character. While it is not yet clear to what extent this lack of agreement as to the optic orientation of the crossites is the result of compositional differences, the latter may well be the operative factor. Willems<sup>4</sup> concludes that the presence of Ca in the constitution of the blue amphiboles has some influence in determining their individual optical properties. It is possible that the rather high proportion of calcium in the constitution of the Anglesey crossite is responsible for its apparently abnormal optical orientation.

#### *Conclusion.*

A more detailed study of the Anglesey 'glaucophane' than has previously been attempted shows that in chemical constitution and in

<sup>1</sup> G. D. Louderback, Bull. Dept. Geol. Univ. California, 1909, vol. 5, p. 360.

<sup>2</sup> G. Murgoci, Compt. Rend. Acad. Sci. Paris, 1922, vol. 175, pp. 426-429. [M.A. 7-221.]

<sup>3</sup> H. A. Brouwer, Bull. Soc. Franç. Min., 1913, vol. 36, pp. 272-275.

<sup>4</sup> H. W. V. Willems, Proc. K. Akad. Wetenschap. Amsterdam, 1937, vol. 40, pp. 720-724. [M.A. 7-250.]

optical properties it has more in common with the crossites than with any other of the amphiboles of the glaucophane-riebeckite series. Although the chemical analysis reveals some 4 % of lime, it is believed that the mineral is best described as a crossite, especially as all analysed crossites show some degree of 'contamination' of this kind.

The writer wishes to record his indebtedness to Professor T. Neville George of the Department of Geology, University of Glasgow, and to Dr. S. R. Nockolds of the Department of Mineralogy and Petrology, University of Cambridge, for reading the script and for helpful suggestions.

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*A note on the occurrence of columbite in the Meldon aplite,  
Devonshire.*

By OLEG VON KNORRING, M.Sc.

Department of Geology, University of Leeds.

[Read November 1, 1951.]

**D**URING a departmental excursion to South Devon in April 1951, material was collected from the Meldon aplite quarry near Okehampton, to the north-west of the Dartmoor granite, which on examination has proved to contain the mineral columbite. The mineral is present only in very small amounts and as extremely minute crystals; nevertheless its presence is of considerable interest in that this appears to constitute the first authenticated record of the occurrence of a mineral of the columbite-tantalite series in Britain.

The following minerals have previously been recorded from the Meldon aplite quarry.<sup>1,2</sup> They are partly found as accessory constituents in the main fine-grained aplite rock consisting of orthoclase, albite, and quartz, or more frequently and in larger amounts in the coarser portion of the aplite and particularly in veins of pegmatitic character.

|          |            |                 |
|----------|------------|-----------------|
| Fluorite | Axinite    | Montmorillonite |
| Quartz   | Tourmaline | Petalite        |
| Apatite  | Cordierite | Orthoclase      |
| Topaz    | Prehnite   | Albite          |
| Garnet   | Lepidolite | Heulandite      |

McLintock<sup>1</sup> has given a detailed account of the mineral assemblages and has described different types of pegmatite veins from the quarry. It would appear that the mineralization has developed in cracks and joints of the aplite, and it should be pointed out that the veins often show a zonal relationship with quartz and mica concentrated along the margins and feldspar in the central portion.

The material in which the columbite occurs was collected from the working quarry on the left-hand side of the entrance road, and consists of a rather distinctive narrow vein of pegmatitic character about 2 cm.

<sup>1</sup> W. F. P. McIntock, On the occurrence of petalite and pneumatolytic apatite in the Meldon aplite, Okehampton, Devonshire. *Min. Mag.*, 1923, vol. 20, pp. 140-150.

<sup>2</sup> Guide to Excursion A.4, Devon and Cornwall (Mineralogical), International Geological Congress, XVIII Session, Great Britain, 1948.

in width, which intersects the normal fine-grained aplite and has the appearance of filling a joint in the latter. The vein tends invariably to split along the centre.

This particular vein contains a considerable amount of a pink mineral, which first attracted attention. However, the mineral did not turn out

to be petalite with montmorillonite, but another alteration product consisting mainly of quartz and a micaceous mineral. McIntock has previously described a similar pegmatite vein and found it to be free of petalite.

The mineral association in this case was as follows: quartz, albite, perthitic feldspar, lithia-mica, green and yellow tourmaline, blue apatite, topaz, fluorite, and tiny black crystals of columbite. A closer examination

FIG. 1. Columbite crystals separated from aplite, Meldon, Devonshire.  $\times 45$ .

of the hand-specimen shows that columbite is not confined to this vein, but is also present in the adjacent fine-grained aplite. A mineral separation was carried out in order to obtain more material for investigation.

The columbite consists of small, often perfectly formed, black and dark-brown crystals up to 0.3 mm. in length. A few of the separated crystals showing the typical thin tabular orthorhombic habit are represented in fig. 1.

The identification was carried out in the first place by X-ray methods, and a powder photograph of columbite from the Meldon aplite is shown in fig. 2, together with powder photographs of columbite from Ivigtut, Greenland, and tantalite from Kigezi, Uganda, for comparison.

A spectrographic determination, made for the writer by Mr. Lars Lund of this department, showed the predominance of niobium and confirmed the identification of the mineral as columbite rather than tantalite.

Minerals of the columbite-tantalite series are frequently found in complex granite-pegmatites and more particularly in those rich in Li, P, and F. The present occurrence is of a similar mineralogical and chemical character, but genetically is probably more closely related to a pneumatolytic phase of crystallization.

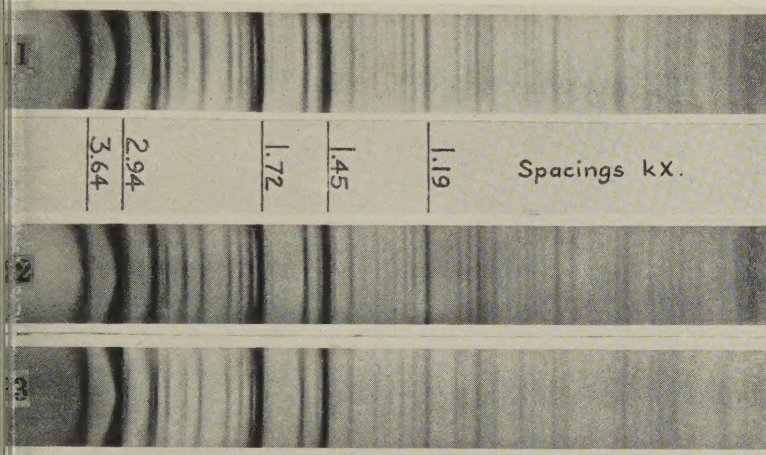


FIG. 2. X-ray photographs of:

- (1) columbite, Meldon, Devonshire.
- (2) columbite, Ivigtut, Greenland.
- (3) tantalite, Kigezi, Uganda.

Cu- $K\alpha$  radiation, camera diameter, 9 cm.





